The Strong Field Simulator: An Attosecond Study of Electron Recollision

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When an atom or molecule is exposed to a strong alternating electric field, such as from a femtosecond pulsed laser, the electron can be tunnel ionized and subsequently driven back to the parent ion in a three-step process called electron recollision. We report on a novel attosecond study of electron recollision using sub-cycle XUV pulses to ionize noble gases dressed by an infrared field of sufficient intensity to drive recollision, this technique is dubbed the Strong Field Simulator. In this experiment, the XUV pulses serve to replace the ionization step while the infrared field still accelerates the electron and drives recollision. Isolating the ionization step from the other two, we can select the moment of ionization, and correspondingly the electron's quantum trajectory, opening new possibilities for studying electron recollision.

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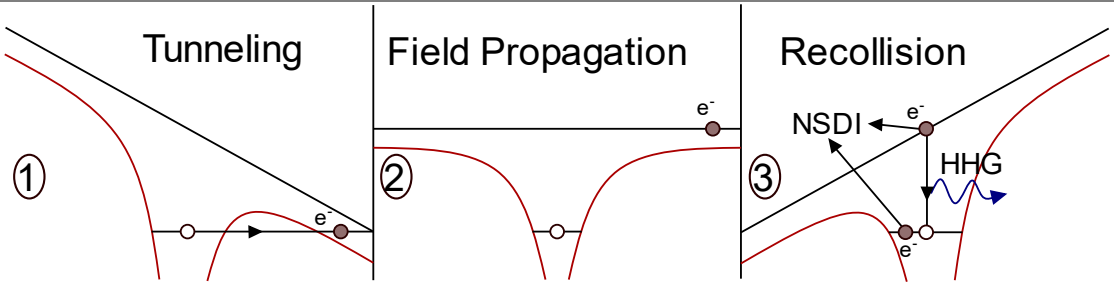
Introduction

An atom or molecule can be ionized by an external electric field if it is strong enough to bend the attractive potential of the nucleus such that the electron can escape via quantum tunneling. This phenomenon has had considerable commercial use in cold cathode fluorescent lighting to backlight LCD monitors and colorful neon lighting for advertising and art. In these cases, a constant electric potential of several thousand volts ionizes gases to form a glowing plasma. However, the alternating electric field of an ultrafast laser pulse, typically femtoseconds (10^{-15}) in duration, can drive this same tunneling ionization process with an interesting twist, whereby the electric field can turn and accelerate the electron to collide with the same atom or

molecule it had escaped [1]. This type of collision between an electron and ion that were previously bound together is called recollision. The electron's trajectory after ionization can be understood classically meaning the probability and energy of the recollision is determined by the moment the electron first escapes the atom or molecule. The three-step process of tunneling ionization, acceleration, and recollision (Fig. 1) has been studied for several decades by measuring the kinetic energies of elastically scattered electrons, the rate of double ionization from inelastic collisions, and the amount of eXtreme UltraViolet (XUV) light produced by recombination of the electron and ion [2].

Figure 1

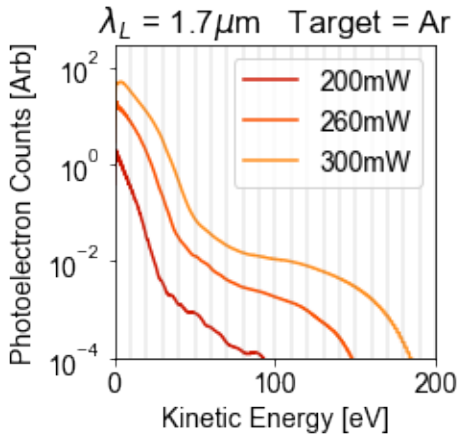
Semi-classical model of strong field recollision



Note. Step 1: The external electric field bends the coulomb potential binding the electron, such that the electron can tunnel into the continuum. Step 2: The free electron is accelerated by the external field. Step 3: The external field drives the electron back into the parent ion resulting in either elastic scattering or inelastic (HHG, NSDI, etc.).

Figure 2 shows a typical energy resolved photoelectron spectrum in a strong field ionization experiment. The large feature at low energy contains electrons that were simply ionized and accelerated by the laser field. The plateau at high energy is associated with recollided electrons (step 3 in Figure 1) that gained additional energy from the scattering process. A figure of merit is the ponderomotive energy $U_p = \frac{e^2E^2}{4m\omega_0^2} \propto I\lambda^2$ where e is the electric charge, E is the electric field amplitude, m is the electron mass, and ω_0 is the carrier frequency of the alternating electric field

Figure 2
Strong field photoelectron spectra

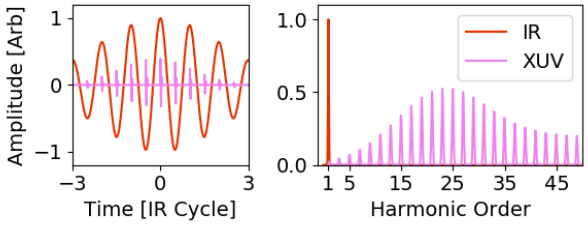


Note. Photoelectron spectra for strong field ionization of Argon with a 1.7μm laser field. The highest electron energy grows linearly with laser power.

[3]. The ponderomotive energy is the classical time averaged kinetic energy of an electron moving in a sinusoidally oscillating electric field. The direct electrons generally peak at kinetic energies of $2U_p$ and the recollided electrons peak at energies of $10U_p$. Figure 2 demonstrates the linear scaling of the ponderomotive energy, as the highest energy electrons are directly proportional to the incident laser power. The quadratic scaling with wavelength is essential to this project as incredibly high ponderomotive energies are attainable with longer wavelength IR sources.

If the accelerated electron is captured by the ion during recollision, then the excess energy is released by emitting a photon. This process is called High Harmonic Generation (HHG) and shown in Figure 4. HHG makes light of sufficient bandwidth to create a train of compressed laser like pulses, ~100 as duration, of XUV radiation (~ 10 – 124 eV). The highest energy photons are equal to 3.17 $U_p + I_p$, where I_p is the ionization energy of the first electron. This has birthed an entirely new field of attosecond (10^{-18}) physics, as the XUV light produced by recollision are laser like pulses of attoseconds duration [4]. For a sense of scale, in one attosecond light will travel the length of a water molecule and in the Bohr model of atomic Hydrogen it takes 152

Figure 3
High harmonic generation



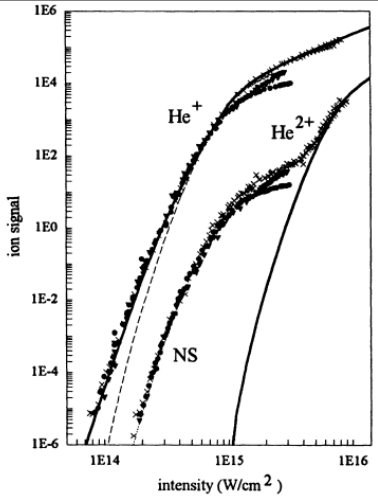
Note. Through HHG, a femtosecond IR laser pulse can be upconverted to a broadband attosecond pulse spanning dozens or even hundreds of harmonic orders.

attoseconds for the lowest energy electron to orbit the nucleus. In this work, I report on a novel attosecond study of recollision where XUV pulses are used to ionize gas targets in the presence of a strong InfraRed (IR) field, this technique dubbed a Strong Field Simulator (SFS).

If the electron recollides with sufficient energy, it is possible to impact ionize the parent ion releasing a second electron in a process called Non-Sequential Double Ionization (NSDI). Figure 4 shows the ionization yield of Helium as a function of laser intensity with wavelength 0.78μm. The solid curve is a prediction from the simple tunneling rate, described by ADK theory, where first one electron then a second is tunnel ionized by the IR field [5]. The measurement features an orders of magnitude enhancement in production of He^{2+} from this prediction which is explained by NSDI. However, below ~$3x10^{14}$ W/cm^2 a recolliding electron has insufficient energy to classically impact ionize the second electron in the ground state. Quantum effects are required to explain NSDI in this region, and a probable culprit is the effects of electron correlation, the quantum description of how electrons interact [5]. Studying NSDI is of particular interest as a fundamental test bed for theories of electron correlation.

Figure 6

Helium ionization rate vs laser intensity



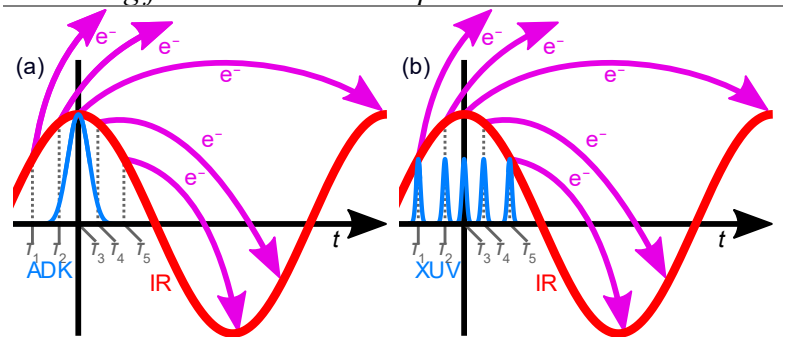
Note. Adapted from [5].

The Strong Field Simulator

One difficulty of studying recollision is that quantum tunneling can happen during a range of times each field cycle, with each time leading to a unique trajectory and recollision energy [3]. Therefore, only a sum of results from many different trajectories are observable, though most electrons ionize near extremes of the laser field as this is when tunneling is most

Figure 5

The strong field simulator concept



Note. A cartoon of the SFS concept. In a typical single color strong field experiment a broad superposition of trajectories are launched, some of which can recollide, with probability proportional to the instantaneous tunneling rate. In the SFS, the attosecond pulses of ionizing radiation select the trajectory launched with fixed probability.

probable, see Figure 5a. Without some additional stimulus, the dynamics of individual trajectories are hidden away from typical strong field experiments. In SFS experiments, we replace tunneling ionization with single photon ionization (step 1), while the IR field still accelerates the electron and drives recollision steps 2 and 3. This allows for control over the ionization time, and therefore the electron trajectory. Separating the ionization step from the IR field, we can isolate and therefore select the moment of ionization, and correspondingly the electron's trajectory, by varying the arrival time of the pulsed XUV and IR fields, see Figure 4b. We observe the outcomes of these singular trajectories by measuring the electron energy spectra in addition to the relative rates of single/double ionization using time of flight spectrometry. The XUV/IR delay, IR and XUV wavelengths, IR intensity, and target species are all controllable parameters allowing for a robust multi-dimensional study of electron recollision.

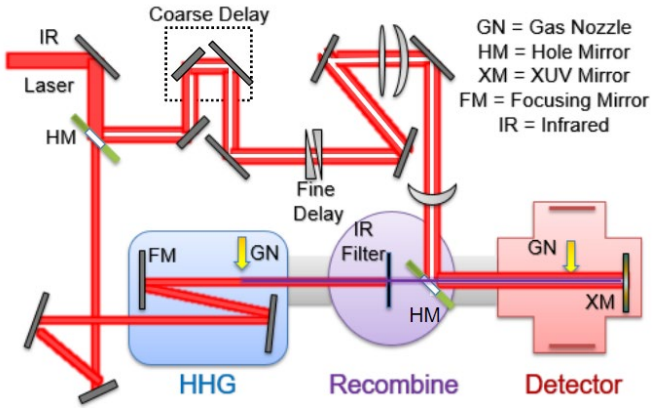
Methods

Attosecond pulses of XUV light are combined with an IR laser in a noble gas target. Controlling the delay between arrival times of the IR and XUV pulses allows for selection of the quantum trajectory after ionization. To ensure sub-cycle control of the delay, a holey mirror splits a single IR laser pulse in two creating an interferometer [4]. In one arm, the IR is focused by a 250mm focal length concave mirror into the output of a gas nozzle which introduces an

atomic vapor into the vacuum system . As the IR focuses into the plume of gas it drives HHG to create the attosecond pulse train of XUV light. This arm of the interferometer is placed in high vacuum to prevent attenuation of the XUV light. A thin (typically 200nm) metallic filter of Aluminum is used to separate the XUV from residual IR light used in the HHG process. The other arm of the interferometer is kept in air, where a pair of glass wedges provide fine control over the pulse delay and a lens assembly matches the divergence of the IR and XUV light. The two beams are recombined using a second holey mirror and both are focused by a dielectric mirror optimized for XUV wavelengths into the target gas inside our detector. Using time-of-flight spectrometry we can simultaneously measure electron kinetic energies and ion charge-to-mass ratio to determine the ionized species.

Figure 6

High harmonic generation



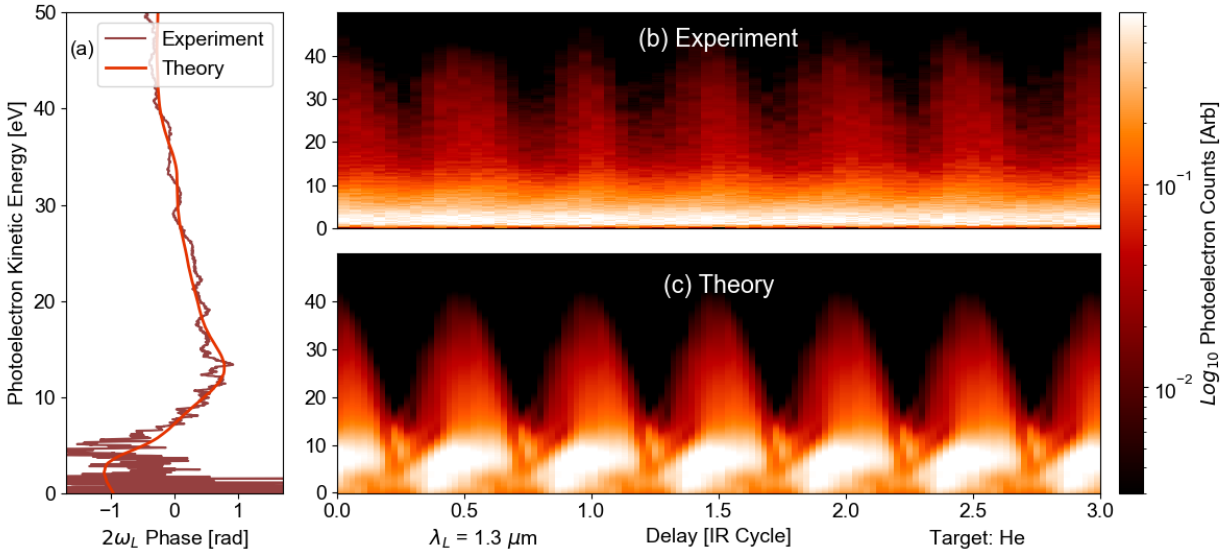
Note. A schematic of a SFS experimental apparatus. The purple beam represents the product of HHG.

The laser system is a Spitfire Ace PA from Spectra-Physics, which is a Ti:Sapphire solid state laser emitting 60fs pulses of 800nm light. The repetition rate is 1kHz with a pulse energy of 12.2 mJ. Using a non-linear process called optical parametric amplification, we can down convert this light to any wavelength between 1200-2100nm. This does reduce the pulse energy to 2.5 mJ but nominally maintains the same pulse duration. The HHG process has an efficiency of 10^{-6}, so XUV pulses of 1 nJ are typical [3]. The aluminum filter has a transmission window in the XUV region from 20-80 eV, which limits the range of photon energies that make it to the interaction region in our detector.

Early Results

A first generation of experiments was conducted at wavelengths of 0.8μm and 1.3μm in a general attosecond interferometer (Fig. 7). In this campaign we could not observe seeded recollision in Helium, so only direct electrons were studied. However, these data validated the SFS concept as an experimental technique [6]. The clearest feature in our experimental data (Fig. 7b) is the oscillation of the highest energy electrons. This can be understood as an electron streaking effect, where electrons are accelerated or decelerated by the IR field depending on the ionization time of the electron. This oscillation occurs twice per IR cycle as HHG produces two attosecond pulses per IR cycle. The phase extracted from a Fourier Filter at twice the laser frequency was in excellent agreement with solutions of the Time-Dependent Schrodinger Equation (TDSE) as calculated by J. Bækhøj (Fig. 7a).

Figure 7
Early results from a first generation of SFS experiments



Note. (b) Photoelectron signal from our first SFS experiments as a function of delay. (c) Solution of TDSE using approximate experimental conditions. Both show a clear oscillation with a frequency of twice per IR cycle. Background ionization by the IR field produce non-oscillating signal at low energies in (b) obscuring the structure seen in (c). The phase of the oscillation demonstrates excellent agreement between theory and experiment in (a). Adapted from [6].

A challenge for the SFS is the photoelectron counts from the IR field tunnel ionizing electrons, both from the desired species and from contaminants in the vacuum chamber. This competing process creates a background signal that does not oscillate as seen in Fig. 7b. This was unavoidable as the IR intensities needed to drive NSDI at the wavelengths studied will also tunnel ionize Helium. Recall that the ponderomotive energy is the key parameter in strong field experiments, and this quantity scales linearly with intensity but quadratically in wavelength. At the ponderomotive energies of interest for the wavelengths used, the required IR intensity will drive significant tunneling ionization. Our solution is to use longer wavelengths, where we can generate the same ponderomotive energies with significantly less IR intensity. This will reduce the background ionization rate and improve our signal to noise in future experiments.

New Apparatus

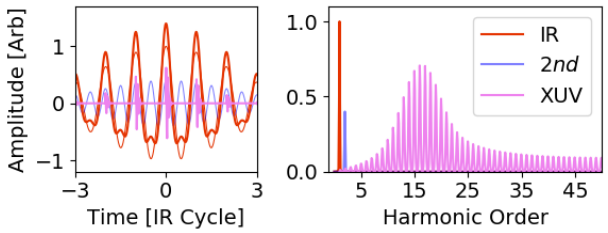
Given the limitations of the first campaign, many of which were constraints imposed by the experimental apparatus, we decided to construct a new attosecond interferometer tailored to performing SFS experiment. Key updates include a dielectric focusing mirror for XUV wavelengths allowing improved bandwidth selection, a tighter focus in the interaction region for increased infrared intensity, a pulsed gas nozzle for higher density targets for HHG [7], an all reflective XUV arm for two color HHG, use of longer wavelength infrared sources, a laser pointing stabilization system, and the use of Xenon gas for improved XUV yield from HHG. With these improvements we believe the next generation of experiments will observe NSDI in Helium.

Two key improvements should be highlighted as they fundamentally change the data collected, as opposed to simply increasing the signal to noise ratio. First, mixing the IR laser with its 2nd harmonic for HHG, or two-color HHG, leads to a fundamentally different attosecond

pulse train (Fig. 8). Adding the second color breaks the symmetry of the electric field, resulting in HHG creating even and odd order harmonics. Correspondingly, there will only be a single XUV pulse per IR cycle, as opposed to the two seen previously. This will double the range of delays observable in the SFS, allowing us to initiate and measure a larger variety of trajectories. Second, adding a dielectric mirror with narrow bandwidth (dot-dashed curve in Fig. 9) provides XUV energy selectivity. Narrowing incident photon energies will limit the number of different trajectories launched by the XUV pulse in each experiment, providing greater resolution for the trajectories of interest. Figure 10 shows the results of our most recent work in this new apparatus.

Figure 8

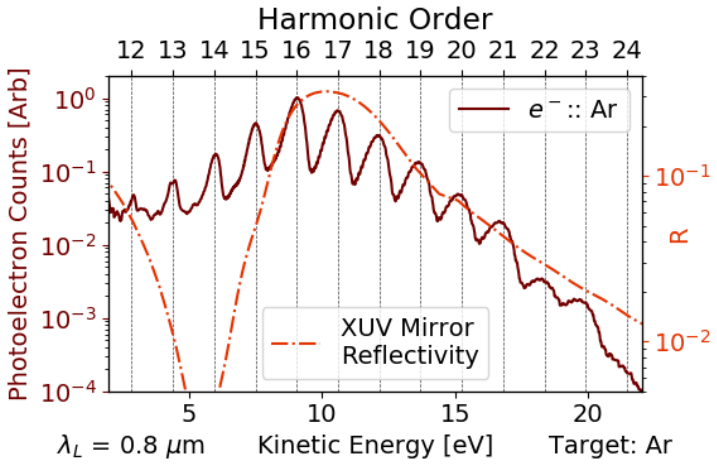
Two-color high harmonic generation



Note. Adding the second harmonic of the IR field to HHG breaks the field symmetry between cycles. This results in the production of even and odd order harmonics, or in the time domain a single attosecond pulse per IR cycle.

Figure 9

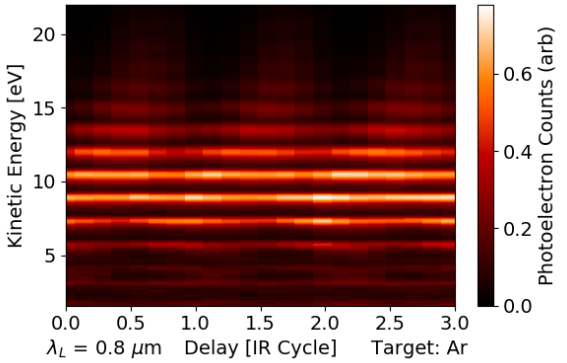
HHG spectrum in new apparatus



Note. This XUV spectrum is measured by single photon ionization of Argon. The dielectric coating effectively reflects a narrow band of XUV light. This mirror's reflectivity is centered at a photon energy of 26eV, with Argon's ionization potential this leads to a peak at electron kinetic energy of 10eV.

Figure 10

Recent experiment in new apparatus



Note. The new apparatus produces very different photoelectron spectrum as opposed to Figure 7. The 2-color HHG leads to a single oscillation per cycle. The narrow bandwidth allows us to clearly resolve both acceleration and deceleration by the IR field.

Conclusion

We have successfully demonstrated the SFS concept, by using attosecond pulses of XUV light to select the ionization time of photoelectrons in the electric field of IR lasers. Changing the delay between the two pulses allows for the selection of specific electron trajectories. With the recent development of a new beamline tailored for SFS experiments, and the use of longer wavelength two color sources, it is anticipated that the next generation of experiments will observe seeded recollision.

This investigation stands to impact the scientific community along two different lines. First, these measurements can inform current efforts to use recollision, and its byproducts, as a means of imaging molecular dynamics with attosecond resolution, the inherent timescale of electron dynamics in atoms and molecules [8]. Second, these experiments provide a fundamental test of electron correlation, the quantum mechanical theory of electron interactions, in the most basic atomic systems. The double ionization in Helium is of particular interest, as electron recollision knocking off a second electron can be thoroughly modeled in this elementary system [9]. Discrepancies between theory and experiment in the distribution of electron energies, the XUV/IR delays maximizing the rate of double ionization, or some other feature will require updating models of electron correlation. These models are fundamental to interpreting and predicting the electronic structure of chemicals and semi-conductors using computational techniques such as Density Functional Theory.

Acknowledgements

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